

# In the United States Patent and Trademark Office

Applicants: Farooq *et al.*

Serial No.: 10/803,749

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For: Oil Containing Starch Granules for  
Delivering Benefit Additives to a Substrate

Examiner: John Hardee

Art Unit: 1751

Confirmation No.: 6908

Attorney Docket No.: 6661-00

## RESPONSE ACCOMPANYING A REQUEST FOR CONTINUED EXAMINATION

In lieu of filing an Appeal Brief in response to the Notice of Appeal filed on July 7, 2006, Applicants are filing a Request for Continued Examination in order to submit additional evidence showing the structure of a starch granule. This response addresses the rejections in the Office Action mailed on June 12, 2006. The claims pending in the application are 1-14.

Note, the Examiner grouped claims 10 and 14 in Group I in the restriction requirement. Claims 10 and 14 are directed to a quaternary ammonium compound, and, so they should be in Group II. Group I should be claims 1, 2, 4, 7, 8, 9, and 11-13. Claims 3, 5, 6, 10, and 14 are correctly indicated as being withdrawn, with traverse.

## OBVIOUSNESS TYPE DOUBLE PATENTING REJECTIONS

Claims 1, 2, 4, 7-9, and 11-13 were provisionally rejected under the judicially created doctrine of obviousness type double patenting as being unpatentable over claims 1-12 of co-pending Application No. 10/803,586. This rejection is rendered moot with the filing of the Terminal Disclaimer on July 7, 2006.

## 35 U.S.C. § 103 REJECTIONS

Claims 1, 2, 4, 7-9, and 11-13 were rejected under 35 U.S.C. § 103(a) as being unpatentable over United States Patent Application Publication No. 2003/0045447 to Heibel et al. in view of United States Patent Application Publication No. 2005/0240670 to Caswell et al.

The claims are directed to a starch granule that has a perfume oil and an organic compound for inhibiting the migration of the perfume oil to the surface of the granule. The perfume oil and the organic compound are absorbed into the granule. The organic compound inhibits the migration of the perfume oil to the surface. By inhibiting the migration, the perfume oil will be available for a longer period of time. This gives a longer lasting fragrance to

a substrate onto which the granule is placed. This is a different solution to the issue of providing a fragrance to a substrate for a longer period of time.

A granule of starch has the structure described below. Neither Heibel '447 nor Caswell '670 disclose granules of starch. The structure in Heibel '447 is a different structure. The structure is a microcapsule that encapsulates material. The material is released when the microcapsule is ruptured. (Abstract) While the microcapsule can be made from polymers, such as polymeric starches, the structure is a microcapsule for encapsulating material. The microcapsule acts as a layer to hold the material until it is released rather than releasing material by de-absorption. There is no disclosure or suggestion in Heibel '447 or Caswell '670 of providing a starch granule with the perfume oil and the compound for inhibiting the migration of the perfume oil to the surface of the granule.

A starch granule is made of layers upon layers upon layers of starch material. From the surface of the particle to the inner part, starch layers are present. This is similar to rings in a tree in that material is layered upon previous material. Enclosed are two web page print outs ([cheng.cam.ac.uk/research/groups/polymer/RMP/nitin/Starchstructure.html](http://cheng.cam.ac.uk/research/groups/polymer/RMP/nitin/Starchstructure.html) and [cermav.cnrs.fr/glyco3d/lessons/starch/page.php.50.html](http://cermav.cnrs.fr/glyco3d/lessons/starch/page.php.50.html)) that show the structure of starch granules.

Attention is directed to page 1 of the [cheng.cam.ac.uk](http://cheng.cam.ac.uk) web page. In the pictures in the lower left corner, an actual starch granule is shown in cutaway so that the layers in the granule can be seen. Figure 3 of the same web page also shows that a starch granule is composed of growth rings. Starch layers are present throughout the granule. These structures are similar to the growth rings in a tree.

Similarly, in the [cermav.cnrs.fr](http://cermav.cnrs.fr) web page, a starch granule is described as having growth rings or "shells", which is shown in the figure. While the term "shell" is used in this document, the "shell" is a growth ring in the granule.

A microcapsule has a different structure. The encapsulated material is contained within a layer of encapsulating material. The encapsulating material does not form layers within the microcapsule. In Heibel '447, it is disclosed that micro encapsulation occurs by polymerizing a polymer shell around droplets or particles (paragraph [0037]). The encapsulating polymer is the outer layer, and the encapsulated material is the inner layer.

Even if a starch polymer is used as the encapsulating material, it will only form an outer layer in the microcapsule. A starch granule structure is not formed by a micro encapsulation process.

From above, it has been shown that starch granules and microcapsules have different structures. As a result of the different structures, the function of each is different. The starch granule with its layers of starch absorbs the perfume oil that is released by de-absorption. Microcapsules release the encapsulated material when the microcapsule is ruptured. The function of a microcapsule does not disclose or suggest the function of de-absorption that is provided by starch granules.

Heibel '447 and Caswell '670 do not disclose or suggest starch granules or the function provided for by starch granules. Therefore, it is respectfully submitted that claims 1, 2, 4, 7-9, and 11-13 are patentable over U.S. Patent Application Publication No. 2003/0045447 to Heibel *et al.* in view of U.S. Patent Application Publication No. 2005/0240670 to Caswell *et al.*

In view of the remarks contained above, Applicants respectfully request reconsideration of the application, withdrawal of the 35 U.S.C. § 103 and obviousness type double patenting rejections, request that all claims be rejoined under MPEP 821.04, and request that a Formal Notice of Allowance be issued for claims 1-14. Should the Examiner have any questions about the above remarks, the undersigned attorney would welcome a telephone call.

Respectfully submitted,

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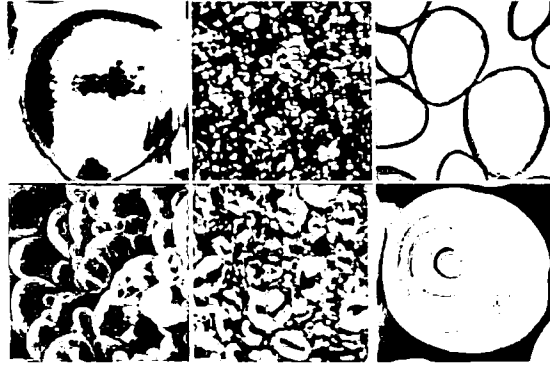
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MFM/dlh

## Starch structure



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## Composition and structure

Starch is produced as granules in most plants cells and is referred to as ?native? when in this particular granular state. Native starches from different botanical sources vary widely in structure and composition, but all granules consist of two major molecular components, amylose (20-30%) and amylopectin (70-80%), both of which are polymers of  $\alpha$ -D-glucose units in the  $^4C_1$  conformation. In amylose (Figure 1), these are linked  $-(1 \rightarrow 4)-$ , with the ring oxygen atoms all on the same side, whereas in amylopectin about one residue in every twenty is also linked  $-(1 \rightarrow 6)-$  forming branch-points as shown in Figure 2.

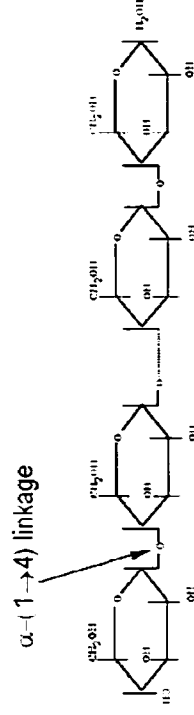


Figure 1 - Amylose molecule

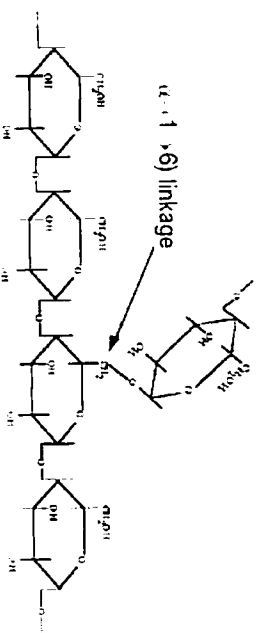


Figure 2 - Amylopectin molecule

Several investigations have been carried out to establish the level of organization within the starch granules. Techniques used vary from X-ray diffraction experiments to atomic force microscopy (AFM) and transmission electron microscopy (TEM). In the native form of starch, amylose and amylopectin molecules are organised in granules as alternating semi-crystalline and amorphous layers that form growth rings as illustrated in *Figure 3*. The semi-crystalline layer consists of ordered regions composed of double helices formed by short amylopectin branches, most of which are further ordered into crystalline structures known as the crystalline lamellae. The amorphous regions of the semi-crystalline layers and the amorphous layers are composed of amylose and non-ordered amylopectin branches.

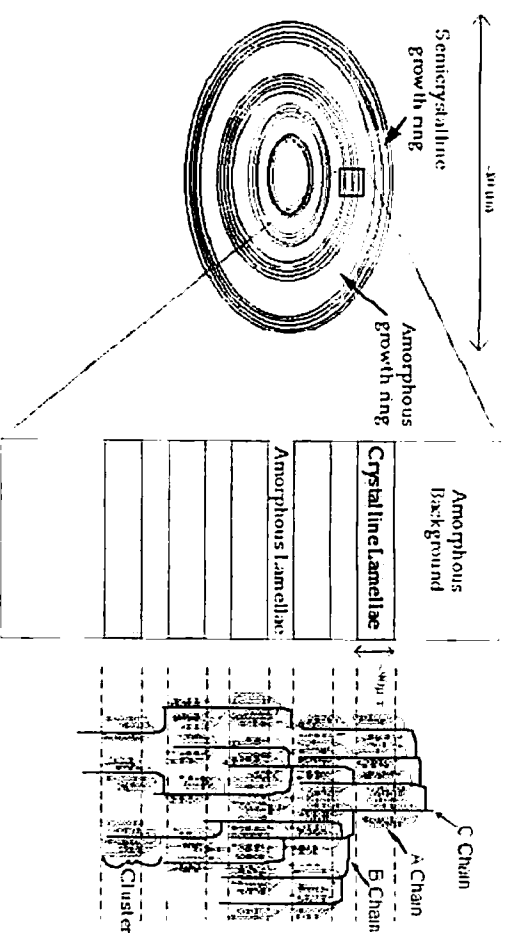


Figure 3 - Schematic view of the structure of a starch granule, with alternating amorphous and semi-crystalline zones constituting the growth rings

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## A NEW HYPOTHESIS OF STARCH GRANULE STRUCTURE - BLOCKLETS (2)

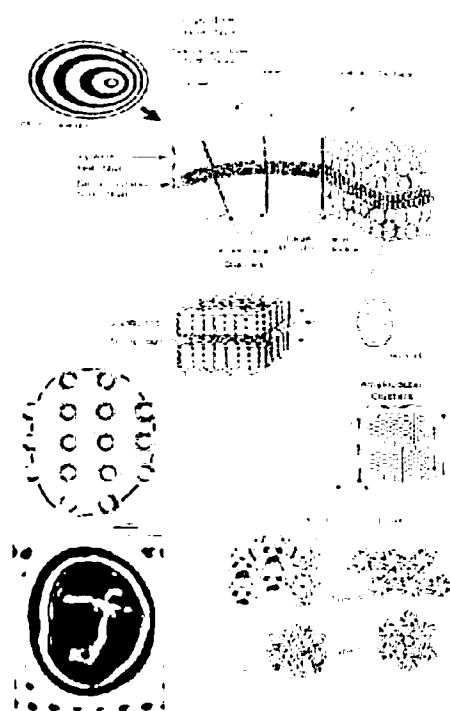
Bertoft <sup>1,2</sup> has demonstrated that the molecules of amylopectin from large and small barley starch granules and of waxy maize starch granules are composed of "super-clusters" with molecular weights of approximately 100 000. These he interpreted as arising from "highly ordered regions of amylopectin". Furthermore, specific (1-4)- $\alpha$ -D-glucosidic linkages between the "super-clusters" appeared to undergo preferential degradation during the initial stages of  $\alpha$ -amylolysis thus producing these "super-clusters". Such observations fit with the idea that an extra level of amylopectin crystallization exists in starch granules and it has been hypothesized by Gallant et al. <sup>1,2</sup> that the "super-clusters" relate to the blocklet structure of starch. The preferentially degraded (1-4)- $\alpha$ -D-glycosidic linkages between the "super-clusters" must therefore be located in the more-amorphous "channel" regions between the blocklets, and are thus more readily accessible to degradation.

Other approaches to structural determination of starch have been performed by Yamaguchi et al. <sup>1,2</sup> and by Oostergetel & van Bruggen <sup>1,2</sup>. These approaches used negative staining of crushed lintnerized starch granules. Yamaguchi et al. <sup>1,2</sup> described some "worm-like ripple structures" in corn starch granules which they interpreted as 5 nm thick crystalline lamellae created by the association of double helices perpendicular to the plane of the lamella. Oostergetel & van Bruggen with a more sophisticated procedure studied lintnerized wheat <sup>1,2</sup> and potato starch <sup>1,2</sup>. Three-dimensional reconstructions of the residual crystallites in potato starch were carried out using negatives taken from a tilt series in the TEM, treated by a low-pass Fourier filter. A helical structure in 3D was observed in stereo mounts, which revealed that the organization of the lamellae was much more complex than was previously thought to be the case. As a result they proposed the concept of "super-helical" structure, which clearly represents a level of structure between that of stacks of lamellae and the granule "growth" ring.

De-branching studies of starch <sup>1,2</sup> lend further support to the idea that a blocklet level of structure exists. Using H.P.L.C., Hizukuri demonstrated that B chains of amylopectin can participate in more than one crystalline amylopectin side chain cluster. He therefore proposed a revised model of amylopectin structure and classified the B chains according to the number of side chain clusters in which they participate. Thus, B1 chains participate in one cluster, B2 and B3 chains extend into 2 to 3 clusters respectively, while B4 chains link 4 or more clusters. It is therefore evident that B chains may link the amylopectin side chain clusters to form larger crystalline units. Such evidence corresponds well with the blocklet concept of starch structure. Furthermore, Hizukuri <sup>1,2</sup> demonstrated that the connecting B chains were more abundant in potato starch and proposed that they are probably characteristic of starches with the B crystal pattern, since such starches have higher amounts of the B2-B4 chain fractions. This observation fits with the observation of Gallant et al. <sup>1,2</sup> that, in general, blocklet size is larger in starches with the B crystal pattern..

The evidence to date in favour of the blocklet concept of starch granule, therefore indicates that the amylopectin lamellae are organized into effectively spherical "blocklets" which range in diameter between 20 to 500 nm depending on starch botanical type and their location in the granule. This organisation fits with the current knowledge of starch granule structure, and is represented schematically in Fig. 22. In general, the "blocklets" are larger (400 to 500 nm in diameter) in starches of the B (e.g. potato starch) and C crystalline type than in starches with the A crystalline type (e.g. wheat starch, in which the "blocklets" were 25 to 100 nm in diameter). In potato starch however, large blocklets (400 to 500 nm in diameter) appear to predominate near the granule surface (approximately the outer 10 $\mu$ m), with smaller blocklets being found nearer the granule centre. In wheat starches, larger (100 nm) "blocklets" are found in the hard crystalline "shells" of the granule than in the softer amorphous "shells" of the granule (blocklet size is around 25 nm in the amorphous shells). Consequently, it has been hypothesized that whilst granule resistance appears to be linked to several interacting factors, the size of the blocklets (i.e. the degree of local crystallinity) may play a rôle in starch granule resistance <sup>1,2</sup>. Blocklet size may therefore play a rôle in the relative resistance of the outer shell of potato starch, and also in the relative resistances of the crystalline and semi-crystalline "growth" shells.

*Overview of starch granule structure (after <sup>1,2</sup> the lowest level of granule organization (upper left), the alternating crystalline (hard) and semi-crystalline (soft) shells are shown (dark and light colours, respectively). The shells are thinner towards the granule exterior (due to increasing surface area to be added to by constant growth rate) and the hilum is shown off centre. At a higher level of structure the blocklet structure is shown association with amorphous radial channels. Blocklet size is smaller in the semi-crystalline than in the crystalline shells. At next highest level of structure one blocklet is shown containing several amorphous crystalline lamellae. In the diagram the starch amylopectin polymer in the lamellae is shown. The next image (from <sup>1,2</sup>) reminds us that amylose-lipid (and protein) features in the organization of the amylopectin chains.*



*the highest level of order, the crystal structure, the starch polymers are shown. (Redraw by [unintelligible] from [unintelligible]).*

The exact location and organisation of amylose in relation to the blocklet organisation is not yet known, however, substantial information regarding the location and state of amylose within the granule is beginning to be gathered and is summarized in section VI below. The presence of locally more-amorphous regions between the blocklets has however been visualised by TEM [unintelligible]. These regions appear to form "amorphous channels" between the blocklets. Whilst it can be hypothesized that a locally higher concentration of amylose (particularly short chains) may be present in such regions, consideration of the ratio of amylopectin to amylose (e.g. 75 to 25 %) in starch granules and the overall crystallinity (15 to 45 %) of starch granules leads to the conclusion that "amorphous" amylopectin is certainly also present in the amorphous regions of the starch granule.

It is therefore evident from the above discussion that considerable evidence exists for a "blocklet" level of granule organization between that of the lamellae and the "growth rings". Clearly, this level of granule structure has significant implications on the internal architecture (and consequently properties) of the starch granule.

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